

Giant Intrinsic Spin-Orbit Coupling in Bilayer Graphene

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The intrinsic spin-orbit coupling (ISOC) in bilayer graphene is investigated. We find that the largest ISOC between π electrons originates from the following hopping processes: a π electron hops to σ orbitals of the other layer and further to π orbitals with the opposite spin through the intra-atomic ISOC. The magnitude of this ISOC is about 0.46meV , 100 times larger than that of the monolayer graphene. The Hamiltonians including this ISOC in both momentum and real spaces are derived. Due to this ISOC, the low-energy states around the Dirac point exhibit a special spin polarization, in which the electron spins are oppositely polarized in the upper and lower layers. This spin polarization state is robust, protected by the time-reversal symmetry. In addition, we provide a hybrid system to select a certain spin polarization state by an electric manipulation.

Recently, ultrathin graphitic devices including monolayer and bilayer graphene have been fabricated experimentally[1, 2]. The experimental developments greatly stimulate theoretical research on this subject[3]. Monolayer graphene is a gapless semiconductor with unique k-linear and massless Dirac-like spectrum, while bilayer graphene presents parabolic and massive spectrum. This distinction in dispersion leads to many physical diversities. To name a few, the Hall plateaus in monolayer graphene appear at half-integer values while that in bilayer graphene appear at integer values without the zero-level plateau[1, 2, 4–6]. The monolayer graphene exhibits the weak anti-localization behavior due to the π Berry's phase, however, in bilayer graphene the 2π Berry's phase leads to weak localization[7]. The difference between monolayer and bilayer graphene has led to great effort in research.

The spin-orbit coupling (SOC) has attracted great attention in the last decade as it plays a very important role in the field of spintronics. SOC can couple the electron spin to its orbital motion and vice versa, thereby giving a useful handle for manipulating the electron spin by external electric fields. Furthermore, the SOC may also generate some exciting phenomena, e.g. the spin Hall effect and the topological insulator.[8] The SOC originates from the relativistic effect, so it is usually very weak. For example, in a monolayer graphene the theoretical research both in tight-binding model and the first principle calculation show that the strength of SOC there is only of the order 10^{-6}eV . [9] It will be very interesting to find a large SOC or to enhance the SOC in a graphene system by some means.

In a carbon atom, the SOC $\Delta\vec{L} \cdot \vec{\sigma}$ (with the angular momentum \vec{L} and spin $\vec{\sigma}$) can couple the $2p_z$ orbit (or π orbit in the graphene) to the opposite-spin $2p_x$ and $2p_y$ orbitals (or σ orbitals in the graphene). The magnitude Δ can be estimated to be of order 5meV . In monolayer graphene, σ orbitals hybridized in a sp^2 configuration to form the honeycomb lattice, while, the remaining $2p_z$ orbit, which is perpendicular to the graphene plane, forms

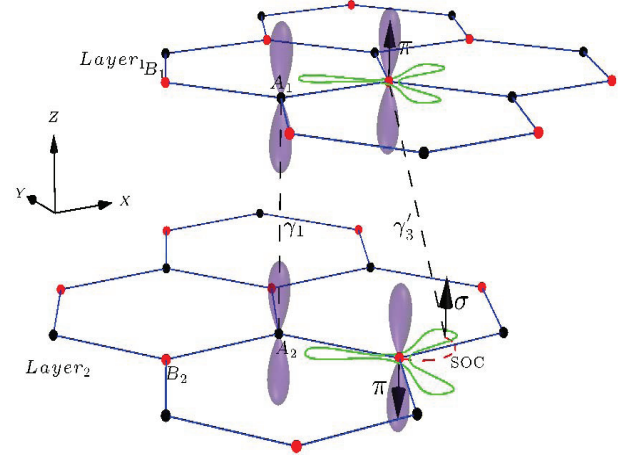


FIG. 1: (color online) Schematic diagram for hopping processes in bilayer graphene, which leads the largest ISOC between π electrons. The black (red) point stands for A (B) sublattice, respectively.

the π orbitals and dominates the low energy physical properties of graphene. Because the π orbitals are perpendicular to the sp^2 orbitals, the hopping process from π orbitals to all σ orbitals are suppressed. Therefore the intrinsic SOC (ISOC) in the low-energy π electrons must involve three hopping events: the SOC between the spin-up π orbital and the spin-down σ orbital, the hopping coupling from the σ orbital to its nearest-neighbor (N.N.) σ orbital with the same spin, and the SOC from the spin-down σ orbital to the spin-up π orbital in the N.N. atom. This ISOC is on the order Δ^2 and its strength is very small, about 10^{-6}eV . [9]. There are several ways proposed to enlarge the SOC strength, [10, 11] e.g. by mixture of π electrons with σ electrons on the N.N. sites by introducing impurities, but this approach will cause the system to be unclean.

In this Letter, we predict a large ISOC in a clean bilayer graphene. We consider bilayer graphene which consists of two coupled honeycomb lattices with sublattices A1, B1 (A2, B2) on the top (bottom) layer, respectively,

arranged by Bernal stacking as illustrated in Fig. 1. Every A1 site lies directly on the top of A2 site, while B1 and B2 do not. We notice the bond of B1-B2 is not perpendicular to sp^2 orbits, so the coupling between the π and σ orbits of the inter-layer B atoms can exist with the strength on the order of 0.1eV . Then as illustrated in Fig. 1, the π electron (red circle) at site B1 can hop to σ orbits (green circle) at site B2 and further hops to the B2's π orbit with the opposite spin through the intra-atom ISOC. This hopping process contributes to the largest ISOC between the π electrons in bilayer graphene, with its strength being on the order of Δ (not Δ^2), so this ISOC should be much large than ISOC in a monolayer graphene.

In order to derive the ISOC between π electrons in bilayer graphene, we need to consider one s orbit and three p orbits for each site, together with two spin indexes, two sub-lattice indexes and two layer indexes, totally there are 32 orbits in every cell. Then, the tight-binding Hamiltonian reads:

$$H = \sum_{\mathbf{i}, \mathbf{j}} \hat{\Psi}_{\mathbf{i}}^\dagger \begin{bmatrix} H_{\sigma, \mathbf{i}\mathbf{j}} & \mathcal{J}_{\mathbf{i}\mathbf{j}} \\ \mathcal{J}_{\mathbf{i}\mathbf{j}}^\dagger & H_{\pi, \mathbf{i}\mathbf{j}} \end{bmatrix} \hat{\Psi}_{\mathbf{j}}, \quad (1)$$

where $\hat{\Psi}_{\mathbf{j}}$ ($\hat{\Psi}_{\mathbf{i}}^\dagger$) is the annihilation (creation) operators of an electron in the cell \mathbf{j} (\mathbf{i}) with its dimension 1×32 (32×1). H_π , H_σ , and \mathcal{J} are the Hamiltonian of π electrons, σ electrons, and the coupling between π and σ electrons. We adapt the parameters of SWM model,[12–14] and the π electron Hamiltonian H_π includes the intra-layer N.N. hopping coupling γ_0 , the interlayer hopping coupling γ_1 of A1-A2 and that γ_3 of B1-B2 (see Fig.1). The value of $\gamma_0 = 3.12\text{eV}$, $\gamma_1 = 0.38\text{eV}$, $\gamma_3 = 0.29\text{eV}$ is adopted from Ref.[12–14]. The on-site energy of π electrons is set at zero as an energy zero-point. For the σ electrons, we choose the atom orbits $2p_x$, $2p_y$, and $2s$ as the base vectors. The on-site energies of Hamiltonian H_σ are 0 for $2p_x$ and $2p_y$ orbits and ϵ_{sp} ($\epsilon_{sp} = -8.87\text{eV}$) for $2s$ orbit. There is no coupling between σ electrons in same atoms because of the orthogonal base vectors. But the hopping couplings between intra-layer N.N. A-B atoms are very strong. We first consider a special case that the bond of A-B atoms is parallel to x axis, thus the hopping Hamiltonian $H_{\sigma h}$ reads:

$$\begin{aligned} H_{\sigma h, AB} &= \hat{\Psi}_{\sigma A}^\dagger \begin{bmatrix} V_{pp\sigma} & 0 & V_{sp} \\ 0 & V_{pp\pi} & 0 \\ V_{sp} & 0 & V_{ss} \end{bmatrix} \hat{\Psi}_{\sigma B} + h.c. \\ &\equiv \hat{\Psi}_{\sigma A}^\dagger V_{hop} \hat{\Psi}_{\sigma B} + h.c., \end{aligned} \quad (2)$$

where $\hat{\Psi}_{\sigma A/B} = (\hat{a}_{2px, A/B}, \hat{a}_{2py, A/B}, \hat{a}_{2s, A/B})^T$ is the annihilation operators of electron in the $2p_x$, $2p_y$, and $2s$ orbits of the A/B atom. In Eq.(2) the B atom must be the N.N. of A. The hopping elements $V_{pp\sigma} = 5.04\text{eV}$, $V_{pp\pi} = \gamma_0 = 3.12\text{eV}$, $V_{sp} = 5.58\text{eV}$, and $V_{ss} = 6.77\text{eV}$. [15] Second, if the bond of A-B atoms

is at an angle θ to the x axis, by taking a rotation in xy plane, the hopping Hamiltonian $H_{\sigma h}$ is obtained as: $H_{\sigma h} = \hat{\Psi}_{\sigma A}^\dagger R^{-1}(\theta) V_{hop} R(\theta) \hat{\Psi}_{\sigma B} + h.c.$, with

$$R(\theta) = \begin{bmatrix} \cos \theta & -\sin \theta & 0 \\ \sin \theta & \cos \theta & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (3)$$

The hopping between σ orbits of inter-layer is neglected because that the σ electron is mainly within each layer.

The π - σ hopping Hamiltonian \mathcal{J} is from both ISOC at each atom and the inter-layer π - σ coupling. The intra-layer π - σ coupling for the N.N. A-B atoms is zero as mentioned above. The inter-layer π - σ coupling in the aligning A1 and A2 atoms is also zero since the A1's π orbit points to the center of the A2 atom. But the inter-layer π - σ coupling between the B1 and B2 atoms exists because that the bond B_1 - B_2 is not perpendicular to the $2p_x$ and $2p_y$ orbits. Such kind of π - σ coupling is a crucial characteristic in bilayer graphene and does not exist in a monolayer graphene. The Hamiltonian of this π - σ coupling is:

$$\mathcal{J}_{\pi\sigma} = \hat{a}_{pz, B1}^\dagger [\gamma'_3 \quad 0] \begin{bmatrix} \cos \phi & -\sin \phi \\ \sin \phi & \cos \phi \end{bmatrix} \begin{bmatrix} \hat{a}_{px, B2} \\ \hat{a}_{py, B2} \end{bmatrix} + h.c., \quad (4)$$

where $\gamma'_3 = \gamma_3 * a/d \approx 0.1\text{eV}$ with the lattice constant a and the distance d between the layers. ϕ is the angle of the projection of the B1-B2 bond in xy plane and the x axis. For every B1 atom there are three N.N. B2 atoms, and their π - σ couplings are similar except for the different ϕ . Similarly, the B2's π orbit also has the coupling with the σ orbits of the B1 atoms. Finally, in the $2s$ and $2p$ base vectors, the SOC $\Delta \vec{L} \cdot \vec{\sigma}$ at each atom site can be easily rewritten as:

$$\mathcal{J}_{SOC} = \frac{\Delta}{2} \hat{a}_{pz\alpha}^\dagger [\alpha \quad i] \begin{bmatrix} \hat{a}_{px\bar{\alpha}} \\ \hat{a}_{py\bar{\alpha}} \end{bmatrix} + h.c., \quad (5)$$

where $\alpha = \uparrow, \downarrow$ is the spin index, $\bar{\alpha} = \downarrow$ (\uparrow) for $\alpha = \uparrow$ (\downarrow), and i is the imaginary unit. Here the SOC is the spin-flip hopping, but all other hoppings mentioned above are between the same spin orbits.

Because that the infinity bilayer graphene is a periodic system, we can rewrite the Hamiltonian in Eq.(1) in the momentum space:

$$\begin{aligned} H &= \int \int (dk_x dk_y / 4\pi^2) \hat{\Psi}_{\mathbf{k}}^\dagger \begin{bmatrix} H_\sigma(\mathbf{k}) & \mathcal{J}(\mathbf{k}) \\ \mathcal{J}^\dagger(\mathbf{k}) & H_\pi(\mathbf{k}) \end{bmatrix} \hat{\Psi}_{\mathbf{k}} \\ &\equiv \int \int (dk_x dk_y / 4\pi^2) \hat{\Psi}_{\mathbf{k}}^\dagger H(\mathbf{k}) \hat{\Psi}_{\mathbf{k}}, \end{aligned} \quad (6)$$

where the momentum $\mathbf{k} = (k_x, k_y)$ is in the first Brillouin Zone.

Next we focus on how to derive the low energy effective Hamiltonian. First the high-energy σ electrons need to be eliminated. For this purpose, we perform a canonical transformation: $\tilde{\mathcal{H}}(\mathbf{k}) = e^{-S} H(\mathbf{k}) e^S$, [9, 16] where

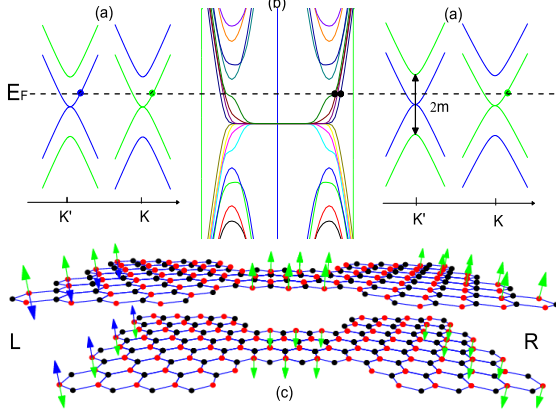


FIG. 2: (Color online) (a) and (b) are the dispersion relations for infinite bilayer graphene (set $k_y = 0$) and zigzag bilayer graphene nanoribbon with width $W = 120\sqrt{3}a$. (c) is the schematic diagram for a hybrid device consisting of two large bilayer graphene connected by a narrow zigzag bilayer graphene ribbon. The green (blue) arrows in (c) represent the spin polarized direction for the green (blue) state in (a).

$S = \begin{pmatrix} 0 & M \\ -M^\dagger & 0 \end{pmatrix}$ is anti-hermitian matrix. After the canonical transformation, we demand that the σ and π electrons are decoupled in the Hamiltonian $\tilde{\mathcal{H}}(\mathbf{k})$. By using the condition of the decoupling and keeping the matrix elements in $\tilde{\mathcal{H}}_\pi$ up to order \mathcal{J}^2 , the matrix M can be obtained:

$$M = -H_\sigma^{-1} \mathcal{J} - H_\sigma^{-2} \mathcal{J} H_\pi - \dots \quad (7)$$

While around Dirac points for the low-energy electron, it is safe to take the approximation $M \approx -H_\sigma^{-1} \mathcal{J}$, because of large energy gap of several eV between σ and π electrons. To substitute this M into the above equation of the canonical transformation, the effective Hamiltonian of π electrons with ISOC reads:

$$\tilde{H}_\pi(\mathbf{k}) = H_\pi - 2\mathcal{J}^\dagger H_\sigma^{-1} \mathcal{J} \quad (8)$$

The magnitude of the ISOC term is roughly estimated as $\Delta\gamma'_3/\epsilon_{\sigma\pi}$, in which $\epsilon_{\sigma\pi}$ is the energy difference between σ and π bonds at Dirac points.

In bilayer graphene, the energy bands of π electron are fourfold except for the spin freedom of degree. Two energy bands touch at Dirac points dominate the low-energy properties, while the other two bands have an energy gap of about $2\gamma_1$ due to the strong interlayer coupling between π electrons on site A1 and A2. Because that the gap $2\gamma_1$ is about 0.7eV , we need to carry out the canonical transformation procedure again to eliminate the high energy π bands for sites A_1 and A_2 . Finally, we get the low-energy effective Hamiltonians H_K

and $H_{K'}$ around the Dirac points K and K' :

$$H_K(\mathbf{k}) = \begin{pmatrix} 0 & Z \\ Z^* & 0 \end{pmatrix} \otimes \mathbf{I}_2 + \begin{pmatrix} 0 & 0 & 0 & m \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ m & 0 & 0 & 0 \end{pmatrix} \quad (9)$$

$$H_{K'}(\mathbf{k}) = \begin{pmatrix} 0 & Z^* \\ Z & 0 \end{pmatrix} \otimes \mathbf{I}_2 + \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & m & 0 \\ 0 & m & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \quad (10)$$

where $Z = \{\frac{v_F^2 k_\pm^2}{\gamma_1} + 3\gamma_3 a k_\pm\}$, $v_F = 3\gamma_0 a/2$ is the Fermi velocity of π electrons, and $k_+ = k_x + ik_y$, $k_- = k_x - ik_y$. In the Hamiltonians H_K or $H_{K'}$, the momentum \mathbf{k} is measured from K or K' , respectively, and the basis orbits are $(p_{z\uparrow}B_1, p_{z\downarrow}B_1, p_{z\uparrow}B_2, p_{z\downarrow}B_2)$. The first term in Eqs.(9) or (10) comes from the well known low-energy Hamiltonian of π electrons on sites B_1 and B_2 of bilayer graphene.[3, 12] The second term is the ISOC between the π electrons, which is a central result of this work. This ISOC origins from the interaction of both the intra-atom ISOC and interlayer hopping that couples the π - σ orbits as shown in Fig.1. Its strength m is about $\Delta\gamma'_3/\epsilon_{\sigma\pi}$, which is on the order of Δ . From our calculation, the strength $m = 0.46\text{meV}$ and it is about 100 times larger than the ISOC in a monolayer graphene.

Eqs (9) and (10) lead to energy bands $E(\vec{k}) = \pm 1/2(|m| \pm \sqrt{m^2 + 4|Z|^2})$. Fig.2a depicts the energy dispersion at fixed $k_y = 0$. Due to the ISOC, the spin degeneracy is removed. Here two bands have an energy gap $2m$ and the two low-energy bands still touch at Dirac points. Around the Dirac point K , the low-energy bands are spin polarized. The spins in the upper layer are mainly upward while they are downward in the lower layer, as shown in Fig.2c. Moreover, this spin polarization is almost independent to the momentum $|\mathbf{k}|$. To compare with the quantum spin Hall effect, in which the spin polarization is opposite on the opposite edges, now the opposite spin polarizations exhibit on the opposite surfaces. Near the Dirac point K' , the spin polarization in the low-energy excitation mode is in contrary to that of K point since they are the time-reversal Krammer pair. Given a time-reversal invariant impurity, the inter-valley scattering processes between these two Krammer's pair are suppressed.[17, 18] Thus, the spin relaxation time is long.

After obtaining the ISOC term in k-space, its tight-binding form in real space can be derived straightforwardly. Here we keep the π orbits of the A1 and A2 atoms, and the tight-binding Hamiltonian is:

$$H = - \sum_{\mathbf{i}, \alpha, \vec{\delta}_i} \gamma_0 [a_{\mathbf{i}\alpha}^\dagger b_{(\mathbf{i}+\vec{\delta}_i)\alpha} + c_{\mathbf{i}\alpha}^\dagger d_{(\mathbf{i}-\vec{\delta}_i)\alpha}] - \sum_{\mathbf{i}, \alpha} \gamma_1 a_{\mathbf{i}\alpha}^\dagger c_{\mathbf{i}\alpha} \\ - \sum_{\mathbf{i}, \alpha, \vec{\delta}_i} \gamma_3 b_{\mathbf{i}\alpha}^\dagger d_{(\mathbf{i}+\vec{\delta}_i)\alpha} + \sum_{\mathbf{i}, \alpha, \vec{\delta}_i} t_{\vec{\delta}_i\alpha} b_{\mathbf{i}\alpha}^\dagger d_{(\mathbf{i}+\vec{\delta}_i)\bar{\alpha}} + h.c(11)$$

where $a_{i\alpha}(a_{i\alpha}^\dagger)$, $b_{i\alpha}(b_{i\alpha}^\dagger)$, $c_{i\alpha}(c_{i\alpha}^\dagger)$, and $d_{i\alpha}(d_{i\alpha}^\dagger)$ denotes the annihilation(creation) operator of the π electron at the A1, B1, A2, and B2 atoms with spin α in the cell \mathbf{i} . $\vec{\delta}_{i=1,2,3}$ are the three N.N. vectors. In equation (11), the first, second, and third terms express the intra-layer and inter-layer π -orbit coupling without spin flip. The last term leads to the spin flip hopping between the B1's and B2's π orbits due to the ISOC, with $t_{\delta_{i\uparrow}} = t_{\delta_{i\downarrow}}^* = \frac{m}{3} \exp(i\vec{K} \cdot \vec{\delta}_i)$. We note that this Hamiltonian maintains C_3 symmetry as well as the time reversal symmetry. By using this tight-binding model, electronic properties of finite bilayer graphene can be calculated.

Until now, we exhibited that the low-energy modes in bilayer graphene are spin polarized due to the ISOC. But in the equilibrium both modes around K and K' are occupied and the total spins are unpolarized. For the purpose of selecting a single excitation mode, we need to choose a valley. We design a hybrid system, the so called 'valley filter'[19] consisting of two wide bilayer graphenes connected by a narrow zigzag bilayer graphene nanoribbon (see the sketch map in Fig.2c), to select a single excitation mode. Earlier works[19, 20] on monolayer graphene ribbon with zigzag edges have shown that there exists a propagating chiral edge state, and the 'chirality' means that the sign of group velocity determines the valley. The situation is the same for the zigzag bilayer graphene ribbon, as demonstrated from its dispersion relation in Fig.2b. Let us set the Fermi energy as shown in Fig.2a and 2b and apply a positive voltage to the device. There will be two kinds of excitation modes (blue and green arrows in Fig.2c) in the left region. However, the edge states of the central zigzag ribbon can only support the one around K point, thus only one excitation mode (green arrows in Fig.2c) transport to the right region after the selection, which is spin polarized. Protected by time reversal symmetry and the large separation between K and K' , the spin polarization in the right region is difficult to be scattered and can last for a long time. Meanwhile, if we change the direction of current flow, the other kind of excitation mode can present in the left region.

In summary, by considering the hopping coupling between interlayer π - σ orbits and the ISOC in carbon atom, we find a large ISOC between π electrons in bilayer graphene, with its strength being around $0.46meV$, about 100 times larger than that in the monolayer graphene. The Hamiltonians of this ISOC in both momentum and real space are derived. Due to the ISOC, the low-energy electrons around the Dirac point exhibit a special spin polarization, in which the electron spins in upper and lower layers are polarized in opposite direc-

tions. In addition, we propose a hybrid system to select a certain spin polarizing mode by an electric manipulation.

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